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Excitons in Cuprous Oxide Crystals at the Temperature of Liquid Helium (4.2°K)

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Previous papers have described the phenomena observed during absorptic of light in cuprous oxide crystals at low temperatures. At the temperature of liquid nitrogen (T = 77.3°K) a hydrogenlike series of narrow absorption lines was found[1,2] which was interpreted as the optical spectrum of the exciton in the Cu_2O lattice. In thinner plates of cuprous oxide at T = 77.3°K a second hydrogenlike series of absorption lines was found[3] on the shortwave side of the first series. We attributed the origin of the second series also to excitons of a second type in the cuprous oxide crystal.

In the present paper we describe further investigations of the absorption spectrum in cuprous oxide at the temperature of liquid helium. The dispersion of the apparatus used was approximately 7 A/mm in the region of the spectrum around λ = 5800 A, three times greater than that of the spectrograph used earlier.

Table I. Wavelengths of absorption lines of the first series at T = 77.3°K and T = 4.2°K.

Ordinal No. of k line	77,3 °K		4,2 °K	
	Wave-lengths λ_k (A)	Energy (ev)	Wave-lengths λ_k (A)	Energy (ev)
1	6125,3	2,0234	6100,6	2,0316
2	5792,7	2,1396	5775,8	2,1458
3	5756,6	2,1530	5840,3	2,1591
4	5743,8	2,1578	5727,4	2,1640
5	5738,1	2,1599	5721,7	2,1661
6	5734,1	2,1615	5717,6	2,1677
⋮	⋮	⋮	⋮	⋮
∞	5727,4	2,1640	5711,7	2,1699

Upon cooling the cuprous oxide crystal to the temperature of liquid helium we observed the following phenomena in the region of the first hydrogenlike series. When the temperature was lowered to 4.2°K, the width of the absorption lines of the first series decreased, and the absorption lines became considerably narrower than at the temperature of liquid nitrogen (77.3°K). The entire series of lines was further displaced toward the shortwave part of the spectrum.

Table I shows the wavelengths of the absorption lines of the first series at the temperature 4.2°K and 77.3°K. (In Table I of reference 2 an error occurred in converting wave numbers into electron volts; it has been corrected in Table I of the present paper.)

The wave numbers of the absorption lines of the first series at the temperature of liquid helium are in good agreement with the series law of hydrogenlike atoms:

$$\nu_k = \nu_\infty - \frac{B}{k^2} = 17508 - \frac{775}{k^2}; \quad k = 2,\ 3,\ 4, \ldots$$

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The values of the constants ν_∞ and R are given in reciprocal centimeters.

The first term of the series ($k = 1$) is an exception here, as at the temperature of liquid nitrogen.

At the temperature of liquid helium in the absorption spectrum of cuprous oxide crystals we observed seven additional weaker and thinner absorption lines, between the lines of the first series. The frequencies of the new lines are shown in Table II.

Table II. Position of the new absorption lines in Cu_2O at $T = 4.2°K$.

Ordinal No.	Wave-lengths λ_k (A)	Energy (ev)	Wave numbers ν_k (cm^{-1})
1	5762,5	2,1508	17353,5
2	5758,0	2,1525	17367,1
3	5751,5	2,1549	17386,7
4	5737,0	2,1604	17430,7
5	5734,5	2,1615	17438,3
6	5731,5	2,1624	17447,4
7	5724,4	2,1651	17469,0

Table III.

Ordinal No.	Wave-lengths λ_k (A)	Energy (ev)	Wave numbers ν_k (cm^{-1})	$\Delta\nu_k$ (cm^{-1}) obs.	$\Delta\nu_k$ (cm^{-1}) calc.
1	—	—	—	—	438,4
2	5751,5	2,1549	17386,7	109,6	109,6
3	5731,5	2,1624	17447,4	48,9	48,7
4	5724,4	2,1651	17469,0	27,3	27,4
.	.	.	.	.	.
.	.	.		.	.
.	.	.		.	.
∞	5715,4	2,1685	17496,3	0	0

Thus, at the temperature of 4.2°K the absorption spectrum of Cu_2O represents a complex system of overlapping groups of narrow lines. It may be supposed that, in addition to the seven lines that we observed, there are still other absorption lines, on which are superimposed broader and more intense terms of the first exciton series, preventing observation of these lines. Graphs *a* and *b* in Fig. 1 show the relative position of the new lines we observed and of the lines of the first series.

Fig. 2 gives photographs of the terms of the first series, between which the weaker lines observed at 4.2°K are visible. The new lines have different absorption coefficients and therefore cannot be brought out with sufficient distinctness simultaneously on the same spectrogram. Fig. 2 therefore shows three spectrograms, *a*, *b*, and *c*, made with different exposures in order to bring out the new lines: 20 min, 5 min, and 5 sec, respectively. The new lines are indicated by marks in Fig. 2.

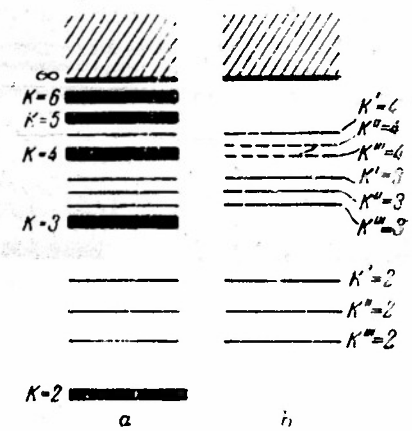

Fig. 1. *a*, relative position of terms of first exciton series (broad lines) and of the new lines in the absorption spectrum of cuprous oxide crystals at $T = 4.2°K$; *b*, series distribution of the new absorption lines in Cu_2O at $T = 4.2°K$. The dashed lines show hypothetical lines.

The characteristic intensity distribution of the terms of the first series is noticeable and is especially marked in the term $k = 2$ in the photograph in Fig. 2 *c*, which was made with the shortest exposure. Here it is plainly visible that the light absorption is distributed very asymmetrically about the line: decreasing gradually on the longwave side, absorption breaks off sharply on the shortwave side.

Among the new lines a group of three lines may be separated out: λ = 5751.5 A, 5731.5 A, 5724.4 A, whose wave numbers, as Table III shows, are in good agreement with the series law of hydrogenlike atoms:

$$\nu'_{k'} = \nu'_{\infty} - \frac{B'}{k'^2} = 17496 - \frac{438.4}{k'^2}; \quad k' = 2, 3, 4\ldots$$

The values of the constants ν'_{∞} and B' are given in reciprocal centimeters.

If we assume the existence of the two lines λ = 5726.3 A, 5729.0 A (shown in Fig. 1 *b* by the dashed lines) which almost coincide with the fourth term of the first exciton series (λ = 5727.4 A) and therefore cannot be observed in the spectrogram, then we can separate out two more groups of lines: λ = 5758.0 A, 5734.5 A, 5726.3 A; and λ = 5762.5 A, 5737.0 A, 5729.0 A. These

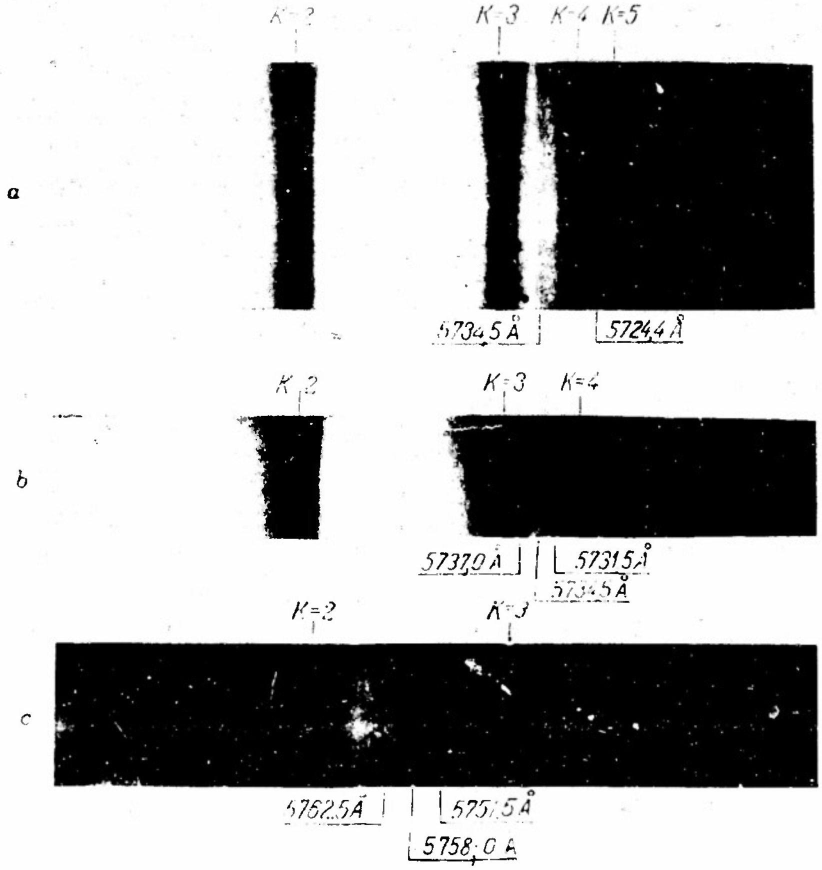

Fig. 2. Spectrograms of the absorption of light in cuprous oxide crystals at temperature of liquid helium (4.2°K) in the region of the first exciton series, made with three different exposures.

two groups also are in agreement with the hydrogenlike series law having wave numbers of the series limits ν''_{∞} = 17,495.2 and ν'''_{∞} = 17,493.1, and the constants B'' = 512 and B''' = 560.

All four series approach the same limit since, within experimental error, the values of ν_{∞}, ν'_{∞}, ν''_{∞}, and ν'''_{∞} coincide.

In addition to the seven new lines described, which are superimposed on the terms of the first series, at T = 4.2°K we found another new limit of continuous absorption (a gradation) at λ = 5841 A (2.1219 ev), and still another very weak line in this gradation, λ = 5817 A (2.1306 ev). We assume that this line is the first term (k = 1) of the second hydrogenlike series.[3]

The fine structure of the first exciton series which we observed shows that the absorption spectrum of cuprous oxide crystals has a very complex character. At the present time it is very difficult to give a reliable interpretation of the new lines. We may indicate here only some possible causes for the appearance of these lines:

(1) Splitting of the exciton levels according to the theory of Davydov.[3,4]

(2) Structure of lines in the exciton spectrum produced by causes of the same kind which produce the appearance of the fine structure of lines in the hydrogen spectrum.

(3) Excitons associated with the presence of impurity atoms and ions in the Cu_2O lattice.

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[1] Ye. F. Gross and N. A. Karryev, Doklady Akad. Nauk SSSR, **84**, 261 (1952).

[2] Ye. F. Gross and N. A. Karryev, Doklady Akad. Nauk SSSR, **84**, 471 (1952).

[3] Ye. F. Gross and B. P. Zakharchenya, Doklady Akad. Nauk SSSR, **90**, 745-48 (1953) [translated in the present series as NSF-tr-122].

[4] A. S. Davydov, *Teoria pogloshchenia sveta v molekulyarnykh kristallkh* [Theory of Absorption of Light in Molecular Crystals] (Kiev, 1951).

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